

Single-spin polaron memory effect

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The single-spin memory effect is considered within a minimal polaron model describing a single-level quantum dot interacting with a vibron and weakly coupled to ferromagnetic leads. We show that in the case of strong electron-vibron and Coulomb interactions the rate of spontaneous quantum switching between two spin states is suppressed at zero bias voltage, but can be tuned through a wide range of finite switching timescales upon changing the bias. We further find that such junctions exhibit hysteretic behavior enabling controlled switching of a spin state. Spin lifetime, current and spin polarization are calculated as a function of the bias voltage by the master equation method. We also propose to use a third tunneling contact to control and readout the spin state.

One of the most promising directions in the fields of molecular electronics and spintronics is the experimental and theoretical investigation of spin manipulation in quantum dots and single molecules. In particular, new methods have been recently developed to investigate spin states of single atoms and molecules using spin-polarized scanning tunneling spectroscopy [1, 2]. Motivated by such achievements the promising question arises whether a single-spin memory effect (including bistability and controlled switching between spin states) is possible.

One of the ways for single spin manipulation is based on the interplay of charge, spin, and vibron degrees of freedom in molecular junctions. In various experiments the signatures of the electron-vibron (e-v) interaction have been observed in atomic scale structures [3, 4, 5, 6]. In the case of strong e-v interaction the formation of a local polaron can lead to a charge-memory effect, which was first predicted long time ago [7], and has been recently reconsidered in more detail [8, 9, 10, 11, 12, 13]. Neutral and charged (polaron) states correspond to different local minima of an effective energy surface and are metastable if the e-v interaction is strong enough. By applying an external voltage, one can change the charge state of this bistable system, an effect that is accompanied by hysteretic charge-voltage and current-voltage curves. A similar memory effect was found in recent STM experiments [14, 15] as a multistability of neutral and charged states of single metallic atoms coupled to a metallic substrate through a thin insulating ionic film; the corresponding polaron model was discussed in [12, 13].

In this Letter we propose an approach to observe a single-spin memory effect by combining the polaron memory mechanism and the spin-dependent tunneling. To this end we consider a single-level and single-vibron quantum system between magnetic leads (Fig.1). We study the case of a symmetric junction with anti-parallel magnetizations of left and right leads, besides the third electrode can be used as a gate or to probe the spin state. The problem can be solved with well controlled approximation in the limit of weak coupling to the leads, where the master equation for sequential tunneling can be used. Thus we focus our major discussion on this limit.

The Hamiltonian of the single-level polaron (Anderson-Holstein) model is

$$\hat{H} = \sum_{\sigma} \tilde{\epsilon}_{\sigma} d_{\sigma}^{\dagger} d_{\sigma} + \omega_0 a^{\dagger} a + \lambda (a^{\dagger} + a) \hat{n} + U \hat{n}_{\uparrow} \hat{n}_{\downarrow} + \sum_{ik\sigma} [(\epsilon_{ik\sigma} + e\varphi_i) c_{ik\sigma}^{\dagger} c_{ik\sigma} + (V_{ik\sigma} c_{ik\sigma}^{\dagger} d_{\sigma} + h.c.)]. \quad (1)$$

Here the first line describes the free electron states with energies $\tilde{\epsilon}_{\sigma}$, the free vibron of frequency ω_0 , the electron-vibron and Coulomb interactions with coupling strength λ and U , respectively; σ is the spin index and $\hat{n}_{\sigma} = d_{\sigma}^{\dagger} d_{\sigma}$, $\hat{n} = \hat{n}_{\uparrow} + \hat{n}_{\downarrow}$. The other terms are the Hamiltonian of the leads and the tunneling coupling ($i = L, R$ is the lead index, k labels the electronic states). The bias voltage V is introduced through the left and right electrical potentials, $V = \varphi_L - \varphi_R$. The energy $\tilde{\epsilon}_{\sigma} = \epsilon_{\sigma} + e\varphi_0$ includes the bare level energies ($\epsilon_{\uparrow} = \epsilon_{\downarrow} = \epsilon_0$ below) and the electrical potential φ_0 describing the shift of the central level by the gate voltage V_G and the bias voltage drop between the left and right lead: $\varphi_0 = \varphi_R + \eta(\varphi_L - \varphi_R) + \alpha V_G$, where $0 < \eta < 1$ describes the symmetry of the voltage drop across the junction, $\eta = 0.5$ stands for the symmetric case considered below.

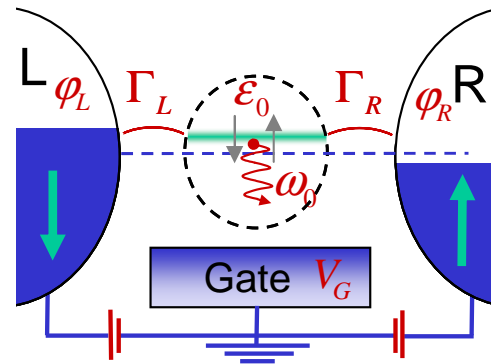


Figure 1: (Color online) Schematic picture of the considered system: a gated single-level quantum dot interacting with a vibron and coupled to ferromagnetic leads.

The coupling to the leads is characterized by the level-width function

$$\Gamma_{i\sigma}(\epsilon) = 2\pi \sum_k |V_{ik\sigma}|^2 \delta(\epsilon - \epsilon_{ik\sigma}). \quad (2)$$

In the wide-band limit considered below, the spin-dependent densities of states in the leads and the tunneling matrix elements are assumed to be energy-independent, so that $\Gamma_{L\sigma}$ and $\Gamma_{R\sigma}$ are constants. The full level broadening is given by the sum $\Gamma_\sigma = \Gamma_{L\sigma} + \Gamma_{R\sigma}$. Below we consider a symmetric junction with antiparallel magnetization of the leads and use the notation $\Gamma_{L\downarrow} = \Gamma_{R\uparrow} = \Gamma$ for majority spins and $\Gamma_{L\uparrow} = \Gamma_{R\downarrow} = \kappa\Gamma$ for minority spins, $\kappa \ll 1$.

The spin effects addressed are particularly pronounced in the limit $U \rightarrow \infty$, i.e. we neglect the doubly occupied state, so that only three states in the charge sector should be considered: neutral $|0\rangle$, charged spin-up $|\uparrow\rangle$ and charged spin-down $|\downarrow\rangle$. Using the polaron (Lang-Firsov) [16, 17, 18] canonical transformation, the eigenstates of the *isolated system* ($\Gamma = 0$) are

$$|\psi_{0q}\rangle = \frac{(a^\dagger)^q}{\sqrt{q!}} |0\rangle, \quad (3)$$

$$|\psi_{\sigma q}\rangle = e^{-\frac{\lambda}{\omega_0}(a^\dagger - a)d_\sigma^\dagger} d_\sigma^\dagger \frac{(a^\dagger)^q}{\sqrt{q!}} |0\rangle, \quad (4)$$

with the eigenenergies

$$E_{0q} = \omega_0 q, \quad E_{\sigma q} = \tilde{\epsilon}'_\sigma + \omega_0 q, \quad \tilde{\epsilon}'_\sigma = \tilde{\epsilon}_\sigma - \frac{\lambda^2}{\omega_0}, \quad (5)$$

where the quantum number q characterizes vibronic eigenstates, which are superpositions of states with different number of bare vibrons.

Taking into account all possible single-electron tunneling processes for both leads, we obtain the incoming and outgoing tunneling rates

$$\begin{aligned} \Gamma_{qq'}^{\sigma 0} &= \sum_{i=L,R} \Gamma_{iqq'}^{\sigma 0} = \sum_{i=L,R} \Gamma_{i\sigma} |M_{qq'}|^2 f_i^0(E_{\sigma q} - E_{0q'}) \\ &= \sum_{i=L,R} \Gamma_{i\sigma} |M_{qq'}|^2 f_i^0(\tilde{\epsilon}'_\sigma + \omega_0(q - q')), \end{aligned} \quad (6)$$

$$\begin{aligned} \Gamma_{qq'}^{0\sigma} &= \sum_{i=L,R} \Gamma_{iqq'}^{0\sigma} = \sum_{i=L,R} \Gamma_{i\sigma} |M_{qq'}|^2 (1 - f_i^0(E_{\sigma q'} - E_{0q})) \\ &= \sum_{i=L,R} \Gamma_{i\sigma} |M_{qq'}|^2 (1 - f_i^0(\tilde{\epsilon}'_\sigma - \omega_0(q - q'))). \end{aligned} \quad (7)$$

Here $f_i^0(\epsilon)$ is the equilibrium Fermi function in the lead shifted by the external potential, $f_i^0(\epsilon) = f^0(\epsilon - e\varphi_i)$, and $M_{qq'}$ is the Franck-Condon matrix element that can be calculated analytically (see Refs. [19, 20, 21, 22, 23] for details of the master equation method and calculation of the tunneling rates). The incoming rate $\Gamma_{qq'}^{\sigma 0}$ describes

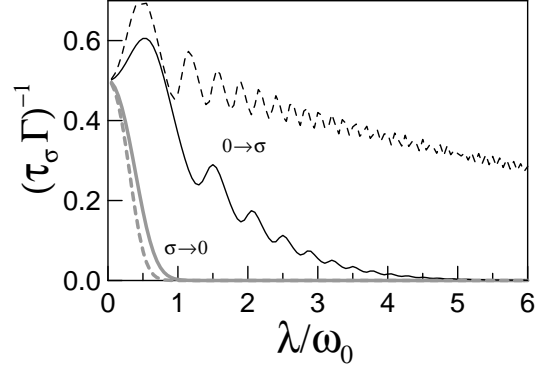


Figure 2: Inverse lifetime $\gamma^{\sigma 0}/\Gamma$ of the neutral state (thin solid line) and the inverse spin lifetime $(\tau_\sigma \Gamma)^{-1}$ (thick gray solid line) as a function of the scaled electron-vibron coupling λ/ω_0 at $\epsilon_0 = \lambda^2/2\omega_0$ and at $\epsilon_0 = 0.1\lambda^2/\omega_0$ (corresponding dashed lines), $T = 0.1\omega_0$.

tunneling of one electron with spin σ from the lead to the dot changing the state of the dot from $|0q'\rangle$ to $|\sigma q\rangle$. The outgoing rate $\Gamma_{qq'}^{0\sigma}$ corresponds to the transition from $|\sigma q'\rangle$ to $|0q\rangle$.

In the sequential tunneling regime the master equation for the probability $P_q^n(t)$, $n = 0, \uparrow, \downarrow$ to find the system in one of the polaron eigenstates (3), (4) can be written as [19, 20, 21, 22, 23]

$$\frac{dP_q^n}{dt} = \sum_{n'q'} \Gamma_{qq'}^{nn'} P_{q'}^{n'} - \sum_{n'q'} \Gamma_{q'q}^{n'n} P_q^n + I^V[P]. \quad (8)$$

Here the first term describes the tunneling transition *into the state* $|nq\rangle$ and the second term the transition *out of the state* $|nq\rangle$. $I^V[P]$ is the vibron scattering integral describing the relaxation of the vibrons to the thermal equilibrium.

Finally, the average charge and the spin polarization are

$$Q = e \sum_q (P_q^\uparrow + P_q^\downarrow), \quad S = \sum_q (P_q^\uparrow - P_q^\downarrow), \quad (9)$$

respectively, and the average current (from the left or right lead) reads

$$J_{i=L,R} = e \sum_{\sigma qq'} (\Gamma_{iqq'}^{\sigma 0} P_{q'}^0 - \Gamma_{iqq'}^{0\sigma} P_q^\sigma). \quad (10)$$

To proceed further, we calculate the characteristic lifetimes of the neutral, spin-up, and spin-down ground states ($q = 0$). We define the switching rates $\gamma^{\sigma 0}$ from the neutral to the charged state with spin σ and vice-versa as the sum of the rates of all possible processes which change these states

$$\gamma^{\sigma 0} = \sum_q \Gamma_{q0}^{\sigma 0}, \quad \gamma^{0\sigma} = \sum_q \Gamma_{q0}^{0\sigma}. \quad (11)$$

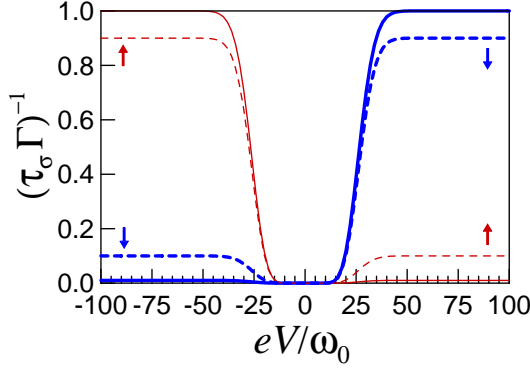


Figure 3: (Color online) Inverse spin lifetime as a function of normalized bias voltage eV/ω_0 at $\lambda/\omega_0 = 3$, $\kappa = 0.01$, $\epsilon_0 = \lambda^2/2\omega_0$ for the spin-up state (thin red solid line) and the spin-down state (thick blue solid line) and the same for a less polarized junction ($\kappa = 0.1$, dashed lines).

In the sequential tunneling approximation the spin lifetime τ_σ is determined by the lifetime of the charged state, from (7), (11). It reads (assuming that the Fermi energy in the leads is zero, see details in [12], $g = (\lambda/\omega_0)^2$)

$$\tau_\sigma^{-1} = \gamma^{0\sigma} = (1 + \kappa)\Gamma \sum_q \frac{e^{-g} g^q}{q!} f^0(-\tilde{\epsilon}'_\sigma + \omega_0 q). \quad (12)$$

At large g the sequential tunneling rates are exponentially suppressed and the cotunneling contribution to τ_σ^{-1} becomes dominant. It can be estimated as [23]

$$\tau_\sigma^{-1(ct)} \approx \frac{\kappa \Gamma^2 T \omega_0^2}{\lambda^4}. \quad (13)$$

Although the cotunneling contribution is not suppressed exponentially by Franck-Condon blockade, it is of the second order in the tunneling coupling and suppressed additionally by the small polarization parameter κ and large λ . At typical parameters, considered in this Letter, the cotunneling contribution can be neglected, but it can be essential at larger tunneling couplings and larger temperatures.

The dependence of τ_σ^{-1} and $\gamma^{0\sigma}$ on the scaled electron-vibron interaction constant $\sqrt{g} = \lambda/\omega_0$ is shown in Fig. 2. For large values of λ the tunneling from the neutral state to the charged state and vice versa is suppressed compared to the bare tunneling rate Γ . Hence all states are (meta)stable at low temperatures and zero voltage. Moreover, the lifetime of the charged states can be much larger than that of the neutral state.

Next we address the other important question, whether fast switching between the two spin states is possible. To this end we consider what happens, if one sweeps the voltage with different velocities, τ_{exp} is the characteristic time of the voltage change. At this point an assumption about the relaxation time τ_V of the vibrons without

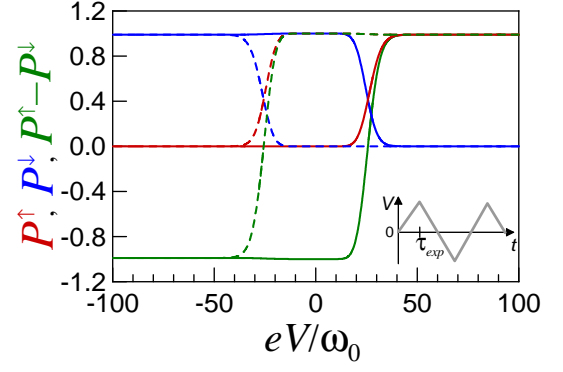


Figure 4: (Color online) Populations of the spin-up state (red), spin-down state (blue), and spin polarization (green) as a function of normalized voltage eV/ω_0 at $\lambda/\omega_0 = 3$ and $\epsilon_0 = \lambda^2/2\omega_0$, the solid (dashed) for increasing (decreasing) voltage. Inset: sketch of voltage time-dependence.

change of the charge state is due. We assume that the relaxation is fast, $\tau_V \ll \tau_\sigma, \tau_{exp}$, so that after an electron tunneling event the system relaxes rapidly into the vibronic ground state $|\sigma 0\rangle$ or $|0 0\rangle$. In this case the probabilities $P^\sigma = \sum_q P_q^\sigma$ of the charged and $P^0 = \sum_q P_q^0$ of the neutral state are determined from the equations

$$\frac{dP^0}{dt} = \sum_\sigma (\gamma^{0\sigma} P^\sigma - \gamma^{\sigma 0} P^0), \quad (14)$$

$$\frac{dP^\sigma}{dt} = \gamma^{\sigma 0} P^0 - \gamma^{0\sigma} P^\sigma, \quad (15)$$

where the switching rates $\gamma^{0\sigma}, \gamma^{\sigma 0}$ at finite voltage are calculated from Eqs. (6,7,11):

$$\gamma^{\sigma 0} = \sum_q \frac{e^{-g} g^q}{q!} [\Gamma_{L\sigma} f^0(\tilde{\epsilon}'_\sigma + \omega_0 q - (1 - \eta)eV) + \Gamma_{R\sigma} f^0(\tilde{\epsilon}'_\sigma + \omega_0 q + \eta eV)], \quad (16)$$

$$\gamma^{0\sigma} = \sum_q \frac{e^{-g} g^q}{q!} [\Gamma_{L\sigma} f^0(-\tilde{\epsilon}'_\sigma + \omega_0 q + (1 - \eta)eV) + \Gamma_{R\sigma} f^0(-\tilde{\epsilon}'_\sigma + \omega_0 q - \eta eV)]. \quad (17)$$

The voltage dependence of the inverse spin lifetime is depicted in Fig. 3. If the voltage is large enough, the Franck-Colomb blockade is overcome and the system is switched into spin-up (spin-down) state at positive (negative) voltage. If the bias voltage is swept fast enough, i.e. faster than the spin lifetime at zero voltage, $\tau_{exp} \ll \tau_\sigma(0)$, both spin states can be considered as stable at zero voltage and hysteresis takes place. This is shown in Fig. 4 where the solid (dashed) lines mark the spin population for increasing (decreasing) bias voltage. In the opposite (adiabatic) limit the voltage change is so slow that the system relaxes into the equilibrium state, and the population-voltage curve is single-valued (Fig. 5).

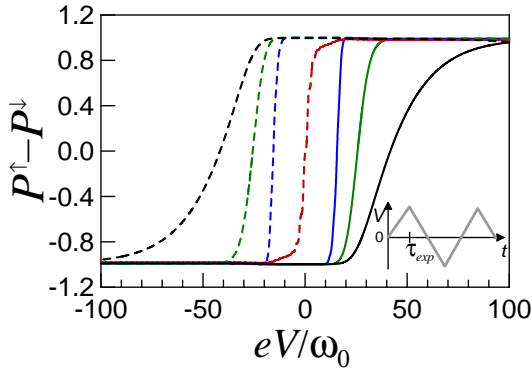


Figure 5: (Color online) Spin polarization as a function of normalized voltage eV/ω_0 at $\lambda/\omega_0 = 3$ and $\epsilon_0 = \lambda^2/2\omega_0$ for three different sweep velocities relative to that in Fig.4 (here shown by green): faster (black), slower (blue) and in the adiabatic limit (red dashed line).

Finally, we study the signatures of the spin polarization in the charge current which is most easily accessible to experiments. In Fig. 6 we show the bias current and the test current to the additional ferromagnetic electrode, very weakly coupled to the system, so that it does not perturb the state. At large negative voltage applied to the electrode the current is sensitive to the orientation of the magnetization in the test electrode, thus the spin state can be controlled during the experiment. Also such a small current can be used to readout the memory element.

In conclusion, we considered a *single-spin memory effect* and switching phenomena in the framework of a single-level quantum dot polaron model, taking into ac-

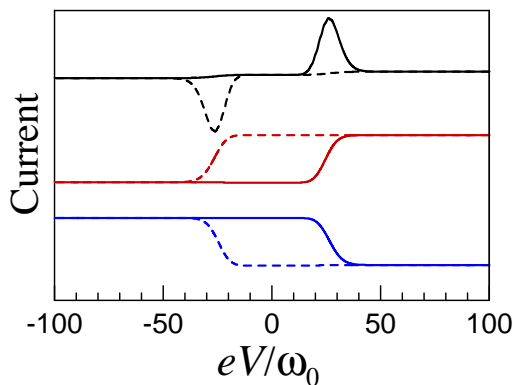


Figure 6: (Color online) Bias current (top black) and test current for spin-up (middle red) and spin-down (bottom blue) test electrode magnetization as a function of normalized voltage eV/ω_0 ; all other parameters are the same as in Fig.4. The curves are shifted along vertical axes and the magnitude of the test current is much smaller than of the bias current.

count non-stationary effects, in particular the interplay between the timescales of voltage sweeping and the quantum switching rates of meta-stable states. We showed that the bistability arises when the quantum switching between two spin states is suppressed due to the Franck-Condon blockade. Controlled switching of the spin can be achieved by applying finite voltage pulses.

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